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Levels and trends of PBDEs and HBCDs in the global environment: status at the end of 2012

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Abstract

In this paper, we have compiled and reviewed the most recent literature, published from January2010 to December 2012, relating to the human exposure, environmental distribution, behaviour, fate and concentration time trends of polybrominated diphenyl ether (PBDE) and hexabromocyclododecane (HBCD) flame retardants, in order to establish their current trends and priorities for future study. Due to the large volume of literature included, we have provided full detail of the reviewed studies as Electronic Supplementary Information and here summarise the most relevant findings. Decreasing time trends for penta-mix PBDE congeners were seen for soils in northern Europe, sewage sludge in Sweden and the USA, carp from a US river, trout from three of the Great Lakes and in Arctic and UK marine mammals and many birds, but increasing time trends continue in polar bears and some birds at high trophic levels in northern Europe. This may be partially a result of the time delay inherent in long-range atmospheric transport processes. In general, concentrations of BDE209 (the major component of the deca-mix PBDE product) are continuing to increase. Of major concern is the possible/likely debromination of the large reservoir of BDE209 in soils and sediments worldwide, to yield lower brominated congeners which are both more mobile and more toxic, and we have compiled the most recent evidence for the occurrence of this degradation process. Numerous studies reported here reinforce the importance o f this future concern. Time trends for HBCDs are mixed, with both increases and decreases evident in different matrices and locations and, notably, with increasing occurrence in birds of prey.

Keywords: PBDEs, HBCDs, distribution, temporal trends, BDE209 debromination

1. Introduction

Since brominated flame retardants (BFRs) were first detected in environmental samples in Sweden in 1979-1981 (Andersson and Blomkvist, 1981) interest in their provenance, environmental distribution and behaviour, fate and effects has grown enormously. A large number of reviews have attempted to document this and to identify directions for future research in order to resolve unanswered questions relating to the significance of the polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs). The first to review this topic was de Wit (2002); amongst the most recent reviews describing environmental levels are de Wit et al. (2010), Dominguez et al. (2012) and Law and Herzke (2012), and analytical aspects have even more recently been reviewed by Dirtu et al. (2013) and Xu et al. (2013). Inputs due to e-waste recycling activities in Asia and their potential effects on the health of local populations have also become of increasing concern over this period (Schnoor, 2012; Zhang et al., 2012a) and it is clear that the international trade in e-waste continues despite regulation (Anon., 2012; BBC, 2010).

In this paper, we aim to compile and assess the most recent evidence (published from January 2010 to December 2012) in relation to the human exposure, environmental distribution, behaviour, fate and concentration time trends of the PBDEs and HBCDs, with a view to identifying research directions and priorities in relation to these well-established BFRs. In view of the sheer volume of the literature reviewed (over 200 papers), we have adopted a novel approach in which this paper provides a summary of the main data reviewed within each topic and details are provided as electronic supplementary information available on the journal website, in order to keep the main paper to a reasonable length. This paper is timely as its data summary bridges the divide between the heyday of the first tranche of flame retardants to be used extensively (the PBDEs and HBCDs) and the rise of their successors, the novel BFRs, which are themselves beginning to be addressed broadly in the literature currently (Covaci et al., 2011; Papachlimitzou et al., 2012; Law et al., 2013).

2. Human exposure

2.1 Indoor air and dust

The low vapour pressures of PBDEs and HBCDs mean that, in indoor environments, they partition preferentially to dust. Consequently, over the period of this review, the majority of studies of indoor contamination address human exposure via ingestion of settled dust, rather than via inhalation of indoor air. Two papers of note have addressed specifically the presence of PBDEs and HBCDs in indoor air over the period of this review. The first reports on the development and validation of a passive air sampler that by providing measurement of both vapour ***and*** particulate phase PBDEs and HBCDs in air, facilitates future monitoring of inhalation exposure to primarily particulate-associated BFRs, like BDE209 and other alternative FRs with similar physico-chemical properties (Abdallah and Harrad, 2010). The second study (Björklund et al., 2012) reported concentrations of BDEs (including BDE209) in air inside 33 buildings to be indistinguishable from those measured simultaneously in air ventilating the same buildings. The authors concluded that contaminated indoor air is an important source of BDEs to

outdoor air. A further study documented concentrations of tri- to hexa-BDEs in both indoor air and dust from 20 indoor locations (homes, offices and laboratories) in Toronto, Canada in 2006 (Zhang et al, 2011a). Although the difference was not significant, the concentrations of ΣBDE_7 in office air (average = 0.79 ng m^{-3}) exceeded those in homes (average = 0.49 ng m^{-3}). These indoor concentrations exceeded substantially those reported for Toronto outdoor air, also suggesting ventilation of indoor air to be a source to the external environment. For PBDEs with 5 or less bromine atoms, significant correlation between air and dust concentrations was observed, confirming the hypothesis that PBDEs in dust arise from gaseous deposition from air. The lack of such correlation for higher brominated BDEs suggests this process to be less important, with other processes such as abrasion of PBDE-containing materials possibly playing a greater role.

Several studies have emerged recently that report levels of PBDEs in dust from countries for which such information has been hitherto unavailable. These studies include reports of concentrations in dust from homes and cars in the Czech Republic (Kalachova et al., 2012), homes and computer suites in Poland (Król et al., 2012), homes in Romania (Dirtu and Covaci, 2010), and offices in South Africa (Kefeni and Okonkwo, 2012). Concentrations of PBDEs in dust from the Eastern European studies were 84 – 5,900 μg Σtri-deca BDEs kg^{-1}, 331-3,100 μg Σtri-hepta BDEs kg^{-1} and 495 μg Σtri-deca BDEs kg^{-1} (median) in Czech, Polish and Romanian homes respectively; thereby providing demonstrable evidence of dust ingestion as an important contributor to the exposure of the population in these countries. HBCDs were also found in Czech and Romanian house dust, at ΣHBCD concentrations of < 0.3 – 950 μg kg^{-1} (Kalachova et al., 2012) and 190 μg kg^{-1} (median) Dirtu and Covaci, 2010), respectively. These concentrations are slightly lower than those reported for countries like the UK and the USA, for which median ΣHBCD concentrations of 730 and 390 μg kg^{-1}, respectively, have been reported previously (Abdallah et al., 2008). The study of Kefeni and Okwonko (2012), revealed median concentrations of BDE47 and BDE99 in office dust from Pretoria, South Africa to be – at 44 and 77 μg kg^{-1}, respectively - in line with those detected in the UK (median values 23 and 65 μg kg^{-1} for BDE47 and BDE99respectively; Harrad et al., 2008) and Japan (median values of 31 and 38 μg kg^{-1} for |BDE47 and BDE99, respectively; Suzuki et al., 2006), but an order of magnitude lower than in the USA (median values of 978 and 1,760 for BDE47 and BDE99, respectively; Batterman et al., 2010). In contrast, levels of BDE209 in South African office dust – at an average of 53 μg kg^{-1} – were substantially lower than reported in other countries (average concentrations of 6,930, 30,000 and 2,400 μg kg^{-1} in office dust from the USA, UK and Japan, respectively). These results imply use of penta-mix PBDE but not deca-mix PBDE formulations in South Africa. Overall, recent data reinforce the position that concentrations in indoor dust (especially of the penta-PBDE congeners) are at least an order of magnitude higher than elsewhere. Likewise, recent evidence underlines that – probably due to the more stringent fire safety regulations in the UK – concentrations of BDE209 in indoor dust in the UK are on a par with those in North America and exceed substantially those elsewhere. Moreover, while regional and national variability exists and concentrations in developing and transition economies are often lower than those in developed regions, the burgeoning database serves to emphasise the ubiquitous indoor presence of BDEs and HBCDs.

2.2 Outdoor air

The growing realisation of the importance of indoor contamination as a source of human exposure has arguably shifted attention away from outdoor air. Notwithstanding this, there remains a strong imperative for studies of the levels of PBDEs and HBCDs in outdoor air, in order to understand better how these BFRs distribute through the environment and enter the food chain. A key concern about PBDEs (a characteristic of POPs generally) is their ability to achieve global distribution, even in locations remote from sources, such as the sparsely populated Tibetan Plateau in two recent studies (Wang et al., 2010; Xiao et al., 2012) and Antarctica (Dickhut et al., 2012).

New data on dust contamination with BDEs has also emerged that augments the global database with respect to levels in hitherto uncharacterised or poorly characterised microenvironment categories. These include: cars, offices, and classrooms in chlld daycare centres and primary schools. Another area for which new information has been generated is the spatial and temporal variability in BDE and HBCD concentrations in dust from the same room. Knowledge of such variability provides important insights into the role of putative sources, and of the uncertainty associated with exposure assessments based on single spot samples of dust taken at one point in time and space. A further potential influence on the concentrations of BDEs in indoor dust samples is the particle size fraction sampled and analysed. It is likely that this issue will form a focus for future research over the coming years, given factors such as the likely dependence of BDEs concentration on particle size and the relative bioavailability of BDEs associated with different particle size fractions. A further area likely to attract increasing attention in coming years is the potential for social inequity in exposure to BDEs and related BFRs. This stems from the hypothesis that lower-income households will contain a disproportionate number of older, deteriorating, or poorly-manufactured PBDE-treated furniture. One study reports worldwide maximum concentrations of BDE47 and BDE99 in dust from low-income homes in California (126,000 and 220,000 $\mu g\ kg^{-1}$, respectively) (Quirós-Alcalá et al, 2011). Also in California, Rose et al (2010) determined concentrations of BDEs in blood plasma from 100 children aged 24-60 months, and reported lower maternal education to be significantly associated with higher concentrations in children. Notwithstanding this evidence, much work remains if this hypothesis of social inequity in exposure is to be tested rigorously. For example, higher concentrations in children in the latter study were associated significantly with factors more likely related to higher socioeconomic status; viz higher consumption of pork and poultry (for penta-mix BDE congeners) and the presence in the home of new furniture and mattresses (for nona- and deca-BDE congeners).

The period covered by this review has seen two notable studies that have examined the relationship between measures of indoor contamination of BDEs and body burdens in individuals in contact with such contamination. In Denmark, Vorkamp et al (2011) studied 43 pregnant women reporting concentrations of some penta-mix BDE congeners in dust to be significantly correlated with those in both matched air and placenta samples, indicating indoor exposures may be an important pathway of exposure for these BDEs, but not BDE209. In Massachussets, USA, Johnson et al (2010) reported similar correlation for penta-mix BDEs in house dust and blood serum of 12 male-female couples; with the low detection frequency of BDE209 in serum preventing evaluation of correlation between dust and serum

levels for this congener. Combined, these studies add weight to the hypothesis that indoor contamination can be an important driver of human body burdens of BFRs.

2.3 Dietary exposure

A number of food basket studies have been performed recently and the exposure of the general population to PBDEs and HBCDs has been estimated. In Sweden, fish and dairy products were the major contributors to PBDE intake (Tornkvist et al., 2011). In the same study, HBCD intake was primarily from fish (65%). In the USA, meat products are a major contributor to PBDE intake (Huwe and West, 2011). HBCD levels were low, consistent with the lesser use of this FR, with the highest levels in canned fish. Eggs are another group of food items which can have a high contribution to the daily intake of PBDEs. Levels of PBDEs and HBCD were high in home-produced eggs in the vicinity of e-waste recycling sites in China (Zheng et al., 2012). HBCD levels were lower than those of BDEs in this study. In comparison with a previous study of home-produced eggs from free range chickens in Belgium, levels of BDEs were greater than those of HBCD and BDE209 was the dominant congener (Covaci et al., 2009). Lower levels were also found in eggs from Canada (Rawn et al., 2011).

Most of the studies have indicated a rather diverse contribution of food items to the dietary exposure, ranging from meat, fish, canned products, and eggs. It also appears that diet becomes an important exposure route only when dust levels of BFRs and thus the contribution of dust to total exposure are low. The importance of each exposure route might be country-specific.

It is important to mention that a number of issues have only very recently been investigated, such as bioaccessibility of ingested food, changes in concentrations and patterns of BFRs during cooking processes (Domingo et al., 2011) and modeling of BFR dietary exposure (Trudell et al., 2011a,b). It is expected that they will continue to grow in importance in the next years.

2.4 Levels in human milk and blood

Overall, 7 papers were identified to report PBDEs and/or HBCD temporal trends in human matrices published since January 2010. Years of collection ranged from 1993 to 2009 with sampling matrices including human blood serum and human breast milk. Results were variable. For PBDEs, decreasing temporal trends or a peak followed by plateau were identified in Australia (Toms et al., 2012), Italy (Alivernini et al., 2011), Germany in Lower Saxony (Hoopmann et al., 2012) and Baden-Würtemberg (Link et al., 2012), and in the Great Lakes Basin of the USA (Turyk et al., 2010). In contrast, PBDE concentrations increased over time in Guinea-Bissau (West Africa) (Linderholm et al., 2010) and Ghana (Asante et al., 2011) (Figure 1). HBCD concentrations, where reported, were also variable and no studies showed a temporal trend (Asante et al., 2011; Toms et al., 2012).

Points of interest in these studies were an increase in BDE209, a congener rarely reported in these papers, in children from Baden-Würtemberg, Germany from 2002-2003 to 2008-2009 (Link et al., 2012); and a shift in congener distribution with an increase in the proportion of BDE153 seen in human samples compared to the previously ubiquitously dominant BDE47 (Linderholm et al., 2010; Turyk et al., 2010; Hoopmann et al., 2012). Reasons for this shift are suggested to be either differences in metabolism of BDE congeners, with BDE153 being more persistent, or to be related to changes in exposure due to the phase out of the penta-mix PBDE product, but not the deca-mix PBDE formulation, during the study period (Turyk et al., 2010; Hoopmann et al., 2012).

Other papers published within this timeframe investigating BFR concentrations in human matrices, but not temporal trends, showed some interesting results. Concentrations of BDEs in serum from children in the U.S. have been positively associated with duration of breast feeding and socio-economic status (Bradman et al., 2012; Stapleton et al., 2012) as well as maternal serum during pregnancy and having no safe places to play (Bradman et al., 2012). In a recent study of umbilical and maternal serum as well as human milk, the congener profile changed over the first month of lactation, however, maternal serum PBDE concentrations did not change significantly (Jakobsson et al., 2012).

3. Sewage sludge and wastewater

In sewage sludge samples from China (Yang et al., 2011), Korea (Hwang et al., 2012) and Italy (Cincinelli et al., 2012), BDE209 dominated the profile. In Chicago, USA, from 1975-2008, penta-PBDE concentrations increased initially and leveled-off around 2000 (production of the penta-mix PBDE product ceased in the USA in 2004) (Hale et al., 2012). Concentrations of BDE209 in biosolids rose from 1995-2008, doubling every 5 years, another indication of the size of the environmental reservoir of BDE209 which has been created.

Sewage sludge and wastewater samples will also be important matrices to examine in the future in order to establish which alternative FR compounds are being used as replacements for PBDEs and HBCD, as they concentrate inputs from diffuse industrial and domestic sources into a point source for release to the environment, which can then readily be sampled.

4. Water and suspended particulates

Few data have been reported recently in these matrices, which is unsurprising as their partitioning and accumulation characteristics make other matrices more attractive for study as seawater concentrations are very low. However, a study of novel brominated and phosphorus flame retardants (PFRs) in the River Aire, UK, has highlighted a type of passive sampling device, the ceramic dosimeter, which may be more widely applicable in the future for flame retardant studies (Cristale et al., 2013).

5. Sediments, plants and soils

5.1 Sediments

Many of the studies reported add to concerns relating to debromination and possible consequent toxic effects from the now considerable reservoir and predominance of BDE209 in sediments across the world due to continued and extensive use of the deca-mix PBDE formulation. Evidence for debromination is outlined in a later section of this paper.

In Taihu Lake in China, Zhou et al. (2012) estimated the inventory of BDE209 to be 26.3 tonnes, a substantial amount, although similar estimates have not been made for other lakes. This gives additional weight to the potential significance of BDE209 debromination in the future. BDE209 dominated the PBDE congener profiles in Taiwan (Jiang et al., 2011) Korea (Hong et al., 2010; Lee et al., 2012; Moon et al., 2007), Indonesia (Ilyas et al., 2011), in the Clyde estuary UK (Vane et al., 2010), Canada (Grant et al., 2011) and in the Scheldt estuary (Van Ael et al., 2012). Significant concentrations of HBCDs were observed in sediments from Tianjin, China (Zhang et al., 2013), Korea (Hong et al., 2010) and Indonesia (Ilyas et al., 2011).

Polychaete worms have been shown to be able to remobilise BFRs buried at 50 cm depth to the surface sediments (Josefsson et al., 2010; Dale and Miller, 2008), so contaminants buried at earlier times can re-enter the environmental cycle by this mechanism. This may also prove to have an impact on the remobilisation of BDE209 degradation products in the future.

5.2 Plants and soils

Plants have been shown to be able to translocate and remobilise PBDEs from soils, although at a lower level than for chlorobiphenyls (CBs) (e.g. Zhao et al., 2012). PBDEs were also more susceptible to metabolism than CBs of the same level of halogen substitution. Wu et al. (2012a) demonstrated isomer-specific HBCD accumulation in maize, and consequent oxidative stress and DNA damage. Li et al. (2011) showed that uptake of HBCDs into plants, and so into the human diet, was limited by adsorption to the soil matrix.

6. Biota

6.1 Terrestrial and aquatic-based mammals

Reflecting the paucity of research characterizing the exposure of biota to environmental contaminants in these ecosystems, only four studies have been published between 2010 and 2012. Novel research was presented on the exposure of bats to PBDEs (Kannan et al., 2010). PBDE-exposure of two species of otters in the UK and North America was also characterized.

PBDEs have now been determined in insectivorous brown bats from the USA (Kannan et al., 2010). BDE99 (41%) and BDE47 (39%) dominated adipose ΣBDE concentrations (118-8,290 μg kg^{-1} lipid weight (lw) and BDE28 showed a greater proportion of the BDE burden in their brains than in adipose tissue.

In England and Wales, concentrations of Σ $PBDE_{26}$ in liver of 30 Eurasian otters were 92 – 19,900 μg kg^{-1} lw. BDE47 dominated the profile (78%) of these otters followed by BDE153 and BDE100 (Walker et al., 2012). In the north-eastern US, the hepatic Σ$PBDE_7$ concentrations (15 – 8,480 mg kg^{-1} lw) of a different species of otter, river otters, were also dominated by BDE47 (62%) and did not show any age- or sex-related differences in concentrations (Stansley et al., 2010). On Vancouver Island, Canada, fecal Σ$PBDE_{12}$ concentrations of river otters were low but significantly higher in otters from Victoria Harbour (0.04 – 2.7 mg kg^{-1} lw) than from Esquimalt Harbour (0.01 – 0.44 mg kg^{-1} lw) or outside of the harbours (0.01 – 1.5 mg kg^{-1} lw) (Guertin et al., 2010).

6.2 Birds' tissues and eggs

There have been approximately 46 studies published between 2010 and 2012 addressing concentrations, temporal trends and spatial patterns, in PBDEs and/or HBCDs in birds. Several studies have reported on the isomeric HBCD pattern in bird tissues, with the dominance of α-HBCD apparent in many but not all birds (e.g., Guerra et al. 2012). The highest concentrations of HBCDs have now been observed in birds of prey, notably in a peregrine falcon egg from Montreal, Canada collected in 2007 (ΣHBCDs 14,600 μg kg^{-1} lw; Guerra et al., 2012), with α-HBCD dominating, and in the muscle of a sparrowhawk collected in 1995 from an undeclared location in the UK (ΣHBCDs 19,000 μg kg^{-1} lw; Leslie et al., 2011), with γ-HBCD dominating. Several important review papers describing concentrations and trends of these particular BFRs in bird tissues have been published during this time period, notably an overall review of concentrations in bird tissues by Chen and Hale (2010), a review of PBDEs, HBCD and novel FRs in birds and other wildlife from China (Wu et al., 2012b) and the Arctic (de Wit et al., 2010,;Letcher et al., 2010), and in specific bird species (Henny et al., 2010).

Several studies have concluded that the geographical patterns and trends of PBDEs and HBCDs measured in the eggs of peregrine falcons (Guerra et al., 2012; de Wit et al., 2010; Chen et al., 2012), ospreys (Henny et al., 2011), herring gulls (Fliedner et al., 2012; Sørmo et al., 2011; Carlsson et al., 2011), and other bird species (Custer et al., 2010; Sun et al., 2012a&b), reflect continental usage patterns, human population densities, and/or introduced legislation governing the manufacturing and usage of these BFRs. Other studies have concluded that sources of exposure to PBDEs as reflected in bird eggs have changed over time (e.g., Crosse et al., 2012).

Global comparisons were made of median and maximal PBDE concentrations in bird tissues (liver, eggs; 2005 – 2010) published between 2010 and 2012. A detailed discussion of the methods used in this analysis is provided in the SI. Median PBDE concentrations of bird tissues were similar regardless of diet, ecosystem or continent, contrasting with continental differences in aquatic birds reported over a longer time period by Chen and Hale (2010). However, a similar significant continental pattern was observed

with maximal PBDE levels of freshwater birds (P = 0.02), all of which were piscivorous or omnivorous with one exception (Morales et al., 2012). Freshwater birds in North America had higher PBDE concentrations (N = 6; median: 6775 μg kg^{-1} lw) than those in Asia (N = 12; median: 2100 μg kg^{-1} lw) or Europe (N = 2; median: 560 μg kg^{-1} lw) (Figure 2). These continental differences remained evident (P = 0.5) following the removal of the particularly high maximal PBDE concentration (70,000 μg kg^{-1} lw) in osprey (Henny et al., 2011). The continental pattern for freshwater birds likely reflects the greater historical usage of PBDEs in North America (Chen and Hale 2010 and references therein). Globally, the analysis also revealed that terrestrial birds had higher maximal PBDE concentrations (N = 8; median: 6600 μg kg^{-1} lw) than freshwater (N = 20; median: 3250 μg kg^{-1} lw) or marine birds (N = 8; median: 318 μg kg^{-1} lw) (P = 0.02), possibly reflecting long-range transport patterns of PBDEs.

6.3 Fish, shellfish and benthic invertebrates

Data were reported for a number of areas/species, including a number of fish species from the Rhone River, the Scheldt estuary, the Bizerte lagoon in Tunisia, rivers in Germany, Norwegian waters and from Brazil. In fish from the Gila River in Arizona, USA, high concentrations of ΣBDE_{50} were reported (up to 12.7 mg kg^{-1} wet weight), amongst the highest in the USA (Echols et al., 2013). Congeners with 2,3,4- or 2,4,5- substitution on one ring were seen to be preferentially metabolised.

Despite some evidence of decreasing trends in some species and locations, elevated concentrations of BDEs in fish are still reported. Shanmuganathan et al. (2011) reported ΣBDE_{11} concentrations of 1.0 – 45 μg kg^{-1} fresh weight in edible fish products from markets in Adelaide, Australia. BDE209 was the dominant congener, followed by BDE47, BDE99 and BDE100.

In China, relatively low levels of HBCD were determined in 12 edible fish species from South China. The concentrations of ΣHBCDs ranged from non-detectable to 0.19 μg kg^{-1} wet weight, with a detection frequency of 70%. This was at the lower end of the concentration range observed globally. Moreover, ΣHBCDs concentrations were higher in both freshwater and seawater farmed fish than in wild marine fish, indicating that human activities probably represent an important input source of HBCD in aquaculture (Meng et al., 2012), although whether via discharges to water or direct usage in their activities is unclear.

Similar low HBCD levels in China were reported by Xia et al, 2011, reporting HBCD levels in two species of marine fish from nine Chinese coastal cities. HBCDs were detected in all samples analyzed, indicating ubiquitous contamination of these compounds in the Chinese coastal environment. The average ΣHBCDs concentration was 3.7 μg kg^{-1} lw (range: 0.57-10 μg kg^{-1} lw), which is lower than seen in many other regions of the world, especially Europe, where HBCDs have been used more intensively than elsewhere. Geographically, the highest HBCD level present in fish was found in Dalian, in northern China, and the lowest occurred in Wenzhou (Xia et al., 2011).

In Japan, similarly low levels of ΣHBCDs were observed in 54 wild and 11 farmed seafood samples from four regions. For the fish classified as *Anguilliformes*, *Perciformes*, *Clupeiformes* and farmed *Salmoniformes*, the median concentrations of ΣHBCDs were 2.1, 0.75, 0.12 and 1.3 μg kg^{-1} ww,

respectively. However, HBCDs were not detected in samples classified as *Crustacea, Mollusca, Pleuronectiformes* and *Scorpaeniformes* or, if detected, the levels were very low (Nakagawa et al, 2010).

6.4 Turtles

Significant knowledge about the PBDE exposure of sea turtles, many of which are threatened or endangered, was gained between 2010 and 2012 (Swarthout et al., 2010; Alava et al., 2011; Malarvannan et al., 2011; Ragland et al., 2011; Stewart et al., 2011). As with birds, maternal transfer of PBDEs to eggs and hatchling sea turtles occurs (Stewart et al., 2011), and maternal diet likely influences PBDE egg concentrations (Alava et al., 2011). BDE47 was usually the dominant PBDE congener (Swarthout et al. 2010), but for several species, PBDE concentrations and profiles differed between migratory and residential turtles (e.g., Ragland et al. 2011). A geographical gradient of increasing ΣBDE egg yolk concentrations for loggerhead turtles occurred from around Florida (< 0.1 – 7.8 ng g^{-1} lw) to North Carolina (0.4 – 37 ng g^{-1} lw) (Alava et al., 2011).

6.5 Marine mammals

Concentrations of PBDEs were determined in a number of cetacean species from Brazilian waters, of particular interest as the majority of data available hitherto are for animals from the Northern Hemisphere (Yogui et al., 2011; Leonel et al., 2012; Dorneles et al., 2010). In 10 cetacean species, concentrations in liver ranged from 3 – 5,960 µg kg^{-1} lw, similar to concentrations seen in Northern Hemisphere dolphins. The highest concentration was seen in a false killer whale which feeds over the continental shelf and, in general, higher concentrations were seen in species feeding in estuarine areas or over the continental shelf than in oceanic species.

Mongillo et al. (2012) used an individual-based modelling approach to predict accumulation of ten PBDE congeners in specific individuals within the southern resident killer whale populations off the west coasts of Canada and the USA. Model predictions for current concentrations agreed well with ΣBDE_{10} concentrations measured in biopsy samples from known individuals. The predicted concentrations over the life-span of individual killer whales were consistent with a doubling time of ca. 3 – 4 years.

In adipose tissue of polar bears from western Hudson Bay, sampled at intervals between 1991 and 2007, concentrations of PBDEs showed significant increases of 13% annually between 1991 and 2007 (McKinney et al., 2010). During the 1990s, α-HBCD was not detected initially, but, following 2000, was detected at concentrations 5- to 10-fold lower than those of the PBDEs, and with a maximum concentration observed in 2003. There were differences in the relative proportions of BDE47 and BDE153, with adult males having lower proportions of BDE47 and higher proportions of BDE153 than adult females and sub-adult bears, which significant statistical correlations suggest reflect dietary differences and the modifying role of lactation on dietary uptake by females and sub-adults (McKinney et al., 2010).

6.6 Food chain studies

Vuorinen et al. (2012) studied biomagnification of PBDEs in Atlantic salmon from its main prey species (sprat and herring) in three areas of the Baltic Sea. BDE accumulation seemed to be highly dependent on both age and fat content, illustrating both the lipophilicity of these contaminants and past accumulation in tissues.

Pectoral muscle of kingfishers (*Alcedo atthis*) and their prey fish species from an electronic recycling site in south China were assessed for the concentrations and potential for biomagnification of PBDEs (17 congeners), DBDPE and BTBPE (Mo et al., 2012); here, we report on the PBDE findings only. In the kingfishers, $\sum BDE_{17}$ levels ranged from 2,030 to 26,400 μg kg^{-1} lw (median: 8,760 μg kg^{-1} lw) which is several times higher than those in the muscles of 7 waterbird species (37-2,300 μg kg^{-1} lw) and 3 terrestrial species (1,000-5,200 μg kg^{-1} lw) from the same e-waste recycling site (Zhang et al., 2011b; Sun et al., 2012a). BDE47 was the predominant congener (~39% contribution) followed by BDE154, BDE153, BDE100 and BDE99 in descending order and contributing ~50% to the $\sum BDE_{17}$ concentrations. The biomagnification factors (BMFs) for all PBDE congeners, except BDE28, were greater than unity, with mean BMFs of 1.4 to 4.5 for the $\sum BDE_{17}$ depending on the fish species, and the authors conclude that these were within the range of most BMFs calculated in other bird studies involving different tissues (Mo et al., 2012).

7. Summary of time trends

7.1 Europe

Schuster et al. (2011) collected background soils from 70 locations along a latitudinal transect between the UK and Norway in 2008, 10 years after their first analysis in 1998 (for a map see Law et al. (2008)). Comparisons between the datasets showed a general decline in PBDE concentrations over time. In sewage sludges from Sweden, Olofsson et al. (2012) reported a decrease (ca. 20% each year) in concentrations of BDE154 and BDE183 (representative of the penta-mix and octa-mix PBDE products, respectively) during 2004-2010, while concentrations of BDE209 (the major component of the deca-mix PBDE product) increased by 16% over the same period. The downward trend for BDE154 seemed to be linear, whilst the upward trend for BDE209 seemed to be increasingly steep.

In a study undertaken in Germany (Esslinger et al., 2011), pooled herring gull eggs from the North and Baltic Sea coasts were analysed for HBCDs. During 1998-2008, all egg pools were dominated by α-HBCD, and temporal trends showed increasing concentrations until 2000 before beginning to decline.

7.2 North America

In archived sewage sludge (biosolids) samples from Chicago, USA, from 1975-2008, penta-PBDE concentrations increased and levelled-off around 2000, while deca-PBDE concentrations rose from 1995-2008, doubling approximately every 5 years (Hale et al., 2012). Congener patterns in contemporary biosolids supported the contention that BDE209 can be debrominated to yield lesser brominated PBDE congeners (BDE206, BDE207 and BDE208, in particular, in this case).

In trout from the Great Lakes sampled during 1980-2009, Crimmins et al. (2012) recorded temporal trends for ΣBDE_5. In Lakes Huron, Michigan and Ontario, decreases in concentration were observed after 2000-2001. In Lakes Erie and Superior, concentrations seem to have stabilised, but not begun to decline significantly.

In general, penta-BDE concentrations in a range of environmental media (air, sediment, wastewater effluent, landfill effluent, aquatic biota and birds) reported from Canada increased until approximately 2000, when levelling off or decreasing trends were observed, consistent with the introduction of control measures (Backus et al., 2010).

Chen et al. (2011) found elevated concentrations of HBCDs in fish from the USA. Carp from the Hyco River, Virginia, exhibited contrasting time trends for PBDEs and HBCD. Between 1999-2002 and 2006-2007, mean ΣHBCDs concentrations rose from 13 to 4,640 μg kg^{-1} lw, whilst mean ΣBDE_9 concentrations fell from 40,700 to 9,140 μg kg^{-1} lw over the same period. This suggests that HBCD usage in the USA may have risen following the termination of PBDE manufacture in the USA in 2004.

7.3 The Arctic and Antarctic

In western Hudson Bay, ΣBDE_7 concentrations in the adipose tissue of polar bears showed significant increases of 13% annually between 1991 and 2007 (McKinney et al., 2010).

Rotander et al. (2012) reported time trends of PBDEs in seven species of marine mammals (long-finned pilot whales, minke whales, fin whales, harbour porpoise, Atlantic white-sided dolphins, ringed and hooded seals) from the Arctic and the North Atlantic taken during 1986-2009. The highest levels were found in samples from the late 1990s-2000. In blubber of Weddell seals from the Antarctic taken in 2006, out of 7 PBDE congeners analyzed, BDE47 was detected in only two animals (Trumble et al., 2012).

Muir and de Wit (2010) reported some evidence that environmental levels of penta-PBDE related congeners were leveling off or beginning to decline in Arctic samples. They examined studies of Arctic char, burbot, lake trout ringed seals, northern fulmar, thick-billed murres, Brünnich's guillemots, ringed seals and beluga and concluded that most studies indicated declining concentrations of BDE47 and BDE99. Most surprising was the predominance of BDE209 in air samples, all of it in the particle phase.

8. E-waste recycling

Over the period covered by this review, concern has grown considerably about the potential for environmental contamination arising from the rudimentary treatment of e-waste (Schnoor, 2012). A

particular focus has been on such activities in China (Chi et al, 2011), with a notable number of studies reporting relatively high PBDE and/or HBCD concentrations in birds from e-waste recycling areas in China (Figure 2) (Liu et al. 2010, Yu et al. 2011, Zhang et al. 2011b, Sun et al. 2012a, b). although recent Chinese government actions to limit the importation of e-waste has resulted in attention switching to other regions such as Africa, Thailand, and Vietnam (Chi et al, 2011; Muenhor et al, 2010; Tue et al, 2010). The beneficial impacts of this Chinese government action are beginning to become evident in that country, with decreasing concentrations reported in some studies (e.g. Fu et al., 2012; Zhang et al., 2012b). Future research in this aspect of the environmental impacts of BFRs is likely to focus on a better characterisation of the impacts on human exposure and health, and on filling the substantial gap in knowledge of the extent of such informal e-waste treatment activities in hitherto under-studied regions such as Africa. Also, further study of the formation and release of brominated and mixed brominated/chlorinated dioxins and furans during these activities and their possible health effects is warranted (Leung et al. 2011; Zhang et al. 2012c&d; Nguyen et al. 2013).

9. Environmental and laboratory evidence for the debromination of BDE209 and transformation of HBCDs

Of major concern is the possible debromination of the large reservoir of BDE209 in soils and sediments worldwide deriving from use of the deca-PBDE product, with the potential to yield large quantities of lower-brominated congeners which are both more mobile and more toxic over a considerable time period. This debromination has been shown to occur in sediments from Lake Thun, Switzerland (Bogdal et al., 2010), for example. In sediment cores from the Clyde estuary, UK, the proportion of nona-BDE congeners (BDE206, BDE207 and BDE208) was greater than reported for the deca-BDE product, and this may also represent debromination of BDE209 (Vane et al., 2010). Similarly, the large contribution of both octa- and nona-BDEs to ΣBDE was indicated as being due to BDE209 debromination. The presence of lower brominated BDE congeners in biota from Korea may also point in this direction, as the penta- and octa-PBDE products were little used in that country (Mo et al., 2012). Li et al (2011) studied BDE209 uptake and degradation in ryegrass, reporting microbial degradation of BDE209 with a higher proportion of di- to hepta-BDE congeners in plant tissues than in soil. Huang et al. (2013) studied degradation of four BDE congeners (BDE28, BDE47, BDE99 and BDE209) in *in vitro* root crude enzyme extracts from maize, ryegrass and pumpkin. Degradation of BDE209 was observed in all three extracts, yielding congeners BDE206, BDE207, BDE208 and BDE183 and lower brominated congeners down to BDE7 (which has a low enthalpy of formation; Zeng et al. (2008)), the major proportion of the transformation occurring within 24h of the start of the experiment. A stepwise loss of bromine was observed for all congeners studied, although the patterns of debrominated products produced differed between plant species. Given the large amount of the deca-mix PBDE product in sediments, it would be very useful to establish whether marine plants, from plankton to seaweeds, have a similar capability to degrade BDE209, and at what rates, and this is a topic for future study.

Wei et al. (2013) examined the photolytic debromination pathways of 13 BDE congeners, including BDE209, BDE208, BDE207, BDE206 and BDE183. The congeners were exposed to sunlight in hexane for

up to 64h. Photolytic degradation of BDE209 yielded 45 congeners, with from nona- to tri-BDE substitution. The environmental significance of this process, given the more limited light penetration in many terrestrial and aquatic environments, is unknown.

As described above, Lee et al. (2012) noted the large contributions of nona- and octa-BDE congeners to the total concentrations in riverine sediments of Korea. The relatively large contribution of tri- to hepta-BDE congeners relative to the limited use of the penta- and octa-mix PBDE products in Korea, and the high concentrations of BDE202 and BDE207, may reflect debromination of BDE209 (Mo et al., 2012).Hale et al. (2012) ascribed higher than expected concentrations of BDE206, BDE207 and BDE208 in US biosolids to debromination of BDE209.

Du et al. (2012) exposed zebrafish to the three individual HBCD isomers. Bioaccumulation was concentration dependent, and accumulation efficiencies, biomagnification factors, and half-lives for α-HBCD were the highest of those for the three HBCD isomers, suggesting that these factors may underlie the predominance of α-HBCD in biota samples. Munschy et al. (2011) exposed juvenile common sole to food spiked with six BDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE209) over a 3 month period, followed by 5 months of depuration. Certain BDE congeners not present in the food were detected in fish tissues: BDE49 and another tetra-BDE congener; a penta-BDE congener; BDE154; BDE183 and another hepta-BDE congener; and BDE202. Sole were able to metabolise BDE209 to yield lower brominated congeners and preferential removal of *meta*-substituted bromines suggested that the deiodinase enzyme systems may play a role in the debromination process.

Echols et al. (2013) determined BDEs in fish (common carp, channel catfish and largemouth bass) taken from the Gila River, USA, in 2003. The presence of BDE155, BDE188 and BDE202 all indicated debromination of BDE209 in these fish. Additionally, congeners with 2,3,4- or 2,4,5- substitution were shown to be vulnerable to selective metabolism in carp.

McKinney et al. (2011) studied biotransformation of BDE209 using an in vitro system based on liver microsomes from various Arctic-feeding marine mammals. Significant depletion of BDE209 was observed, although no evidence of simply debrominated metabolites was observed and the biotransformation products could not be identified.

In harbour porpoises during 1992-2008, Law et al. (2010) noted increases in the proportions of BDE153, BDE154 and BDE100. The authors acknowledged that this may have arisen from changes in sources, but concluded that it was more likely to reflect debromination of higher congeners, such as BDE209, in the environment, either by fish or in bottom-living organisms exposed via sediments and then entering the food chain.

10. Knowledge gaps and recommendations for future research

Following earlier steep rises in concentrations of both PBDEs and HBCD, controls on the use of the penta- and octa-mix PBDE products are beginning to yield declines in concentration in environmental

samples. Major concerns still remain in relation to potential debromination of BDE209 and exposure to both people and animals around e-waste recycling sites. Much knowledge has been gained regarding concentrations of BDEs and HBCDs in birds and other wildlife, with concentrations reported from remote locations globally (e.g., Indian Ocean islands).

There is still a need to assess the time trends of PBDEs and HBCDs in human samples for North America (and Europe, at least for HBCDs). PBDE concentrations have decreased in Europe, but time trends for HBCDs are not so clear. PBDE and HBCD trends in Asia are also unclear.

Exposure of children to BFRs would be better assessed by direct measurements of PBDE and HBCD concentrations in their serum, rather than indirectly through estimation of dietary intake and dust ingestion. Some ethical issues relating to this have yet to be overcome, and the potential utility of non- or minimally invasive measures of body burden, such as hair and nails, should be investigated.

While evidence continues to mount that indoor contamination is an important influence on human body burdens, larger studies that evaluate the role of indoor exposures versus diet would be welcome. Likewise, while indoor dust contamination appears to correlate with body burdens for some individuals; studies that clarify the exact mechanism(s) via which this occurs (e.g. ingestion, dermal contact) are required.

Due to the ongoing restrictions on PBDEs and the impending inclusion of HBCDs in the Stockholm Convention on POPs, there has been a shift in the use of FRs, with the recent increasing consumption of novel BFRs and OPFRs. Investigation of occurrence and time trends in various environmental media, biota and human samples for these alternative FRs is strongly recommended.

There is a gap in knowledge concerning the metabolism/debromination of BDE209 and HBCD in birds, and concerning the potential effects of these FRs and PBDEs in birds and mammals. Also, there is minimal information regarding the exposure and possible effects of these compounds in amphibians and reptiles.

E-waste recycling and dumping sites are a continuing topic of interest for research, especially in Asia and Africa. Further monitoring of human exposure and environmental contamination in these regions are warranted. In particular, the formation and release of brominated and mixed brominated/chlorinated dioxins and furans during these activities and their possible health effects should be addressed.

Given the large amount of the deca-mix PBDE product in sediments, it would be very useful to establish whether marine plants, from plankton to seaweeds, have a similar capability to degrade BDE209 to that of the terrestrial plants mentioned above (see section 9), and at what rates, and this is a topic for future study.

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FIGURE CAPTIONS

Figure 1. Concentration of BDE47 (μg kg^{-1} lw) in human milk by year and country of collection (*results from human blood serum)

Figure 2. Significant continental differences were evident in maximal ΣPBDE concentrations (μg kg^{-1} lw) measured in tissues (liver, eggs) of birds inhabiting freshwater ecosystems (Kruskal Wallis; P = 0.02). Concentrations were higher in freshwater birds from North America than those in birds from Asia or China, even after the removal of the highest levels reported by Henny and his colleagues (2011). Tissues were collected between 2005 and 2010. Only minor inconsistencies are expected to have occurred in the statistical results as a result of the varying number of BDE congeners (11 to 18) used to calculate the ΣBDE levels. Letters below each bar refer to the original study: A – Henny et al., 2011; B – Klosterhaus et al., 2012; C – Mora et al., 2012; D – Chen et al., 2012; E – Gentes et al., 2012; F – Morales et al., 2012; G – Mo et al., 2012; H – Zhang et al., 2011b; I – Wang et al., 2012.